

MEASUREMENT OF THE MASS SPECIFIC EXTINCTION COEFFICIENT FOR ACETYLENE AND ETHENE SMOKE USING THE LARGE AGGLOMERATE OPTICS FACILITY

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The mass specific extinction coefficient, σ_s , of smoke produced from acetylene and ethene fuel burned under laminar and turbulent conditions was measured using the Large Agglomerate Optics Facility. Key design features that enable a threefold reduction in the uncertainty compared with previous measurements include a 10-times longer pathlength, less than 0.05% drift in the light intensity ratio, steady-state smoke generation and dilution, accurate flow calibration, and more precise filter weight measurements. The measurements of σ_s are consistent with previous results obtained for smoke from a variety of fuels for both small- and large-scale fires. Specifically, the σ_s of 7.80 m²/g for acetylene smoke produced by a turbulent flame using the new apparatus, is in excellent agreement with 7.82 m²/g as reported by Choi et al. [3] for the same fuel. However, these values are significantly larger than the value of 4.5 m²/g obtained from the study of Wu et al. [7] for acetylene smoke from turbulent flames. The reliability of the present experimental measurements is supported by an absolute calibration using an aerosol comprised of particles of known size, density, and refractive index. The measured values of σ_s in this study appear to be inconsistent with the values of the refractive index of smoke widely used in the combustion community.

Measurements of the specific extinction coefficient for acetylene and ethene smoke indicate that σ_s depends on fuel type but displays little dependence on flame conditions (laminar or turbulent cases). For ethene smoke, the average specific extinction coefficient is 12% higher than for acetylene smoke. The larger σ_s may be due to a beam-shielding effect that is dependent on the primary particle size and the number of spheres comprising an agglomerate.

Introduction

The mass specific extinction coefficient of smoke, σ_s , is needed for determining the mass concentration of postflame smoke via light extinction measurements. An important application for this nonintrusive method is the determination of smoke yield for materials found in constructed facilities using furniture and cone calorimeters [1]. Time-resolved measurements of smoke concentration are also required for validation of field and zone computational models for smoke flow and dispersion in buildings and in the atmosphere for oil spill fires.

Light extinction measurements are typically used to provide a line of sight average mass concentration of smoke. Bouguer's law as applied to smoke is the basis for relating optical measurement and mass concentration. Specifically, Bouguer's law relates the ratio of the transmitted and incident intensities to the mass concentration of smoke M_s (mass/volume), the path length through the smoke, L , and σ_s via the following expression:

$$\frac{I}{I_0} = \exp(-\sigma_s M_s L) \quad (1)$$

In contrast to the conventional filter collection and gravimetric method, the light extinction technique is nonintrusive, temporally resolved, and convenient to implement.

The general utility of this approach is based on the hypothesis that the σ_s is nearly universal for postflame smoke produced from overventilated fires. The basic qualitative ideas that support this hypothesis are that soot from all flames is basically carbon with primary sphere sizes much smaller than the wavelength of light and a fractal dimension less than 2. For these conditions, the light-absorption cross section is proportional to the mass and is the dominant contribution to the light extinction coefficient. There will be a smaller contribution from the light-scattering cross section that depends on the agglomerate size. A brief review of experimental results relevant to the universality of σ_s will be presented.

TABLE 1
Summary of measurements of specific extinction coefficient at 632.8 nm

Laboratory	Fuels	Scale	Avg. Results m ² /g	SD ^a of Avs. m ² /g	U ^b m ² /g
Factory Mutual: Newman and Steciak [4]	heptane, kerosene, Douglas fir, PMMA, PVC, PC, PS, styrene-butadiene rubber	small to large scale	10.2	0.2	
NIST: Mulholland et al. [1] Dobbins et al. [2]	crude oil, heptane, propane, wood cribs, rigid polyurethane cribs	furniture calorimeter, 50 to 350 kW	8.2	0.4	
Georgia Tech: Patterson et al. [5]	PMMA, HDPE, PVC, PC, PS, PP, wood, kerosene, diesel fuel 2 and 5	small scale—1–5 kW	8.5	1.0	
NIST: Choi et al. [3]	acetylene	McKenna premixed flame	7.8	0.5	1.1
University of Michigan: Wu et al. [7] ^d	propane, ethene, propene, acetylene	5–7 kW	4.6	0.2	0.9
University of Essex: Colbeck et al. [6]	petrol, diesel, fuel oil, paraffin oil, butane	5 mL of fuel	1.4	1.0	

^aSD, standard deviation.

^bU, expanded uncertainty at 95% level of confidence.

^c σ_s , obtained from value reported as the ratio of extinction coefficient to particle volume fraction.

^d σ_s , obtained from reported dimensionless extinction coefficient.

At the National Institute of Standards and Technology (NIST), σ_s was determined for wood, rigid polyurethane, and several hydrocarbon fuels for fires ranging in size from 50 to 350 kW [1]. The light source was a He–Ne laser with a wavelength of 632.8 nm. The mean and standard deviation based on the averages for each of the five different fuel types is 8.2 ± 0.4 m²/g, while the result based on all of the 56 individual tests, 8.3 ± 1.0 m²/g, has about twice the standard deviation.

The results from these NIST tests as well as more recent tests performed at NIST [3] and tests from other laboratories [4–6], all performed at a wavelength of 632 nm, are summarized in Table 1. Each study indicated a relatively small fuel dependence; the ratio of the standard deviation to the average σ_s for each study ranged from a minimum of 2% to a maximum of 12%. However, there is a wide disparity in the fuel-averaged σ_s among the various studies with a ratio of 2.3 for the largest to the smallest value. It should be pointed out that the inferred soot concentrations using the range of reported σ_s values will produce more than a twofold variation. The results by Wu et al. [7] appear to be an outlier relative to the previous values [1–6]. The results of the other five studies are clustered around two values: one value being 8.0 m²/g and the other, 10 m²/g.

For two of the studies [1,3], the expanded uncertainty, U , which corresponds to a 95% level of confidence and is equal to twice the combined standard

uncertainty, has been calculated including uncertainty components that can (type A) and cannot (type B) be treated by statistical methods. Type A and type B correspond to the random and systematic components of uncertainty. Major factors contributing to the uncertainty include the concentration gradient in the direction of laser beam propagation, the small change in intensity ratio for low-concentration smoke, and the uncertainty in the flow through the filter. The value of U obtained by Choi et al. [3] was 14% of the mean, whereas the value obtained by Wu et al. [7] was approximately 20%. In either case, the uncertainties are too large to detect the relatively small changes that are expected for fuel and burner dependence.

In the experiments of Wu et al. [7], the refractive index was calculated based on differential scattering and extinction measurements. Their value of $1.71 + i0.53$ at 632.8 nm was similar to the values reported by Dalzell and Sarofim [8] ($1.56 + i0.46$ for propane soot and $1.57 + i0.50$ for acetylene soot). The agreement between the two measurements of refractive index presented a dilemma, because the measurement of σ_s by Wu et al. [7] appears to be an outlier with respect to previous measurements. The five previous studies listed in Table 1 have consistent values of σ_s , but the corresponding range of refractive indices (based on numerical calculations to be discussed in later sections) is well outside the range of values of the refractive index of soot reported in the

TABLE 2
Specific extinction coefficient of various smokes measured with the LAOF

Fuel Type	Date, # of Tests	Burner, Diameter	Fuel Flow, cm ³ /s	σ_s , m ² /g	ω^a
Acetylene	17 tests on 6 days	laminar, 1 cm	0.80 (44 W)	7.55 \pm 0.05 ^b	0.220 \pm 0.007
	21 tests on 4 days	turbulent, 5 cm	47 (2.6 kW)	7.80 \pm 0.08	0.250 \pm 0.004
Ethene	20 tests on 6 days	laminar, 1 cm	4.8 (270 W)	8.50 \pm 0.11	0.186 \pm 0.007
	19 tests on 3 days	turbulent, 5 cm	36 (2.0 kW)	8.79 \pm 0.28	0.213 \pm 0.010

^a ω is the ratio of the scattering cross section to the extinction cross section. The \pm quantity is the standard deviation of the repeat measurements.

^bFor each day, the mean of the measured values of σ_s is computed. This is done to account for the day-to-day variations in addition to the variation between results within the same day. The \pm quantity is the standard deviation of these means.

literature. The need to resolve this dilemma motivated our development of a technique for a more accurate and reliable measurement of σ_s .

There are three key elements to the present approach. The first element is to develop a facility capable of measuring σ_s with an uncertainty at least a factor of 2 smaller than previous measurements for determining fuel and burner effects. The second element is to perform an absolute calibration of the measurement system by using an aerosol with a known value of σ_s . These measurements represent the first instance in which an absolute calibration is performed for a σ_s measurement. The third element is to perform measurements with the same burner and fuels as in the previous study [7] to facilitate direct comparisons.

Experimental Description and Uncertainty Analysis

Experiments were performed using the Large Agglomerate Optics Facility (LAOF) to accurately measure σ_s of smoke for acetylene and ethene fuels burning under laminar and turbulent conditions (see Table 2). The LAOF was originally developed to study the optical properties of smoke agglomerates as they grew in the optical cell. Figure 1 displays a schematic of the experimental apparatus including the LAOF and the separate laminar and turbulent burner systems. The laminar burner is similar to one used in previous studies by Samson et al. [9] except that the glass tubes have been replaced with brass tubes. The fuel nozzle has an o.d. of 12.7 mm, and the outer brass tube has an o.d. of 10.8 cm. A thread of smoke emitted by a laminar flame is mixed with dilution air as it flows through a tripper plate. A bypass valve (BV 1) is positioned directly before the diluter to purge the transmission cell of all smoke after each experiment. The turbulent burner is 50 mm i.d. and is wrapped with copper tubing for water cooling [10]. The burner is placed below a 25-cm-diameter fume hood as shown in Fig. 1. The distance

between the burner and the fume hood, $H \sim 36$ cm, is adjusted so that the tip of the flame is just below the hood. All of the smoke is collected by the hood with minimal disturbance to the flame. A 9.5-mm-o.d. copper tube is positioned in the middle of the 10-cm-diameter stack. The sampling end of the tube is tapered to an inside diameter of 5 mm for near-isokinetic collection of the soot and gas mixture. The mixture is then diluted with air prior to entrance into the transmission cell. The temperature and pressure for the soot and gas mixture in the transmission cell are monitored for each experiment, and the temperature is typically within 0.2°C of ambient pressure and the pressure within 50 Pa (0.5 cm of water) of ambient pressure.

The soot and gas mixture enters at location 1 and exits at location 2 of the transmission cell. The transmission cell is a fabricated Pyrex tube with dimensions of 10 cm i.d. and a total length between positions 1 and 2 (denoted in Fig. 1) of 1.26 m. Light from a 10-mW, stabilized He-Ne laser is split to provide a reference measurement for the incident light intensity. To prevent soot deposition on the optical surfaces, air-purged "light tubes" are placed at locations in which the laser beam entered and exited the transmission cell. After exiting the transmission cell, the beam is reflected off a 45° mirror and then split to a photodiode to monitor the transmitted intensity and to a beam dump. The acceptance angle of the detector relative to the transmission beam exiting the tube is 1.1°. The relative uncertainty in the measurement of $\ln(I/I_0)$ is reduced from 3.5% for a previous study [3] to a value of 1.5% using this transmission system with a background drift of I/I_0 on the order of 0.03% over a 20-min period. By increasing the path length L from 0.084 to 1 m, the relative uncertainty in the path length associated with the purge flow is reduced from 2.4 to 0.5%.

A brief overview of the measurement procedure is now given. Once a steadily burning flame is established, the intensity ratio of the transmitted and incident photodiode is monitored for approximately

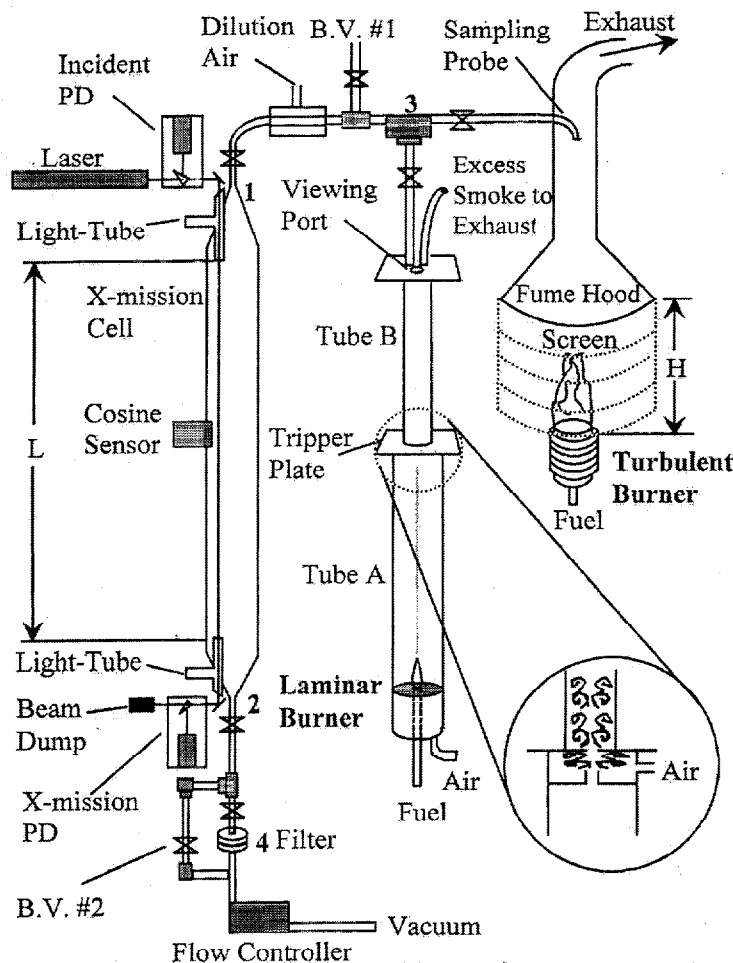


FIG. 1. Schematic of Large Agglomerate Optical Facility and laminar and turbulent burners. An inset displays the tripper plates and dilution air ports to enhance mixing in the laminar burner apparatus.

60 s (denoted by A in Fig. 2). After the background measurement, soot is introduced to the transmission cell by closing BV 1 but leaving BV 2 open (denoted by B). When the intensity ratio stabilizes at a value typically in the range 10–15% below the background value (case with no soot), the exhaust flow is directed through the filter (by closing BV 2) to collect the soot (denoted by C). The soot collection time is approximately 5 min (however, the actual duration is defined by the sooting propensity for the operating condition). After completing the soot collection, both bypass valves are opened to allow clean air to pass through the cell (denoted by D). At a nominal flow rate of $167 \text{ cm}^3/\text{s}$ (10 L/min), the cell is purged in ~ 60 s. The intensity measurements for the background are again recorded to ascertain whether soot was deposited on the optical windows during the collection period (denoted by E). Filters are weighed

before and after each experiment to determine the amount of collected soot. An electrostatic neutralizer consisting of a small α emitter ($500 \mu\text{C}$, Po^{210}) was attached to the top of the balance housing to neutralize the highly charged Teflon filters. The σ_s is then calculated using equation 1.

Soot is sampled using a stainless steel filter housing with Teflon filters [1,3,7]. The filter has a collection efficiency greater than 99% efficiency for particles larger than $0.1 \mu\text{m}$. There was no evidence of soot penetrating the First of a pair of filters. The improvements in the mass concentration measurement include using a balance with a factor of 5 lower uncertainty (with repeatability of 2–3 μg) and an automated soap film flowmeter with a relative uncertainty of 0.9% compared with 2% in the previous study [3].

The filter collection and gravimetric technique

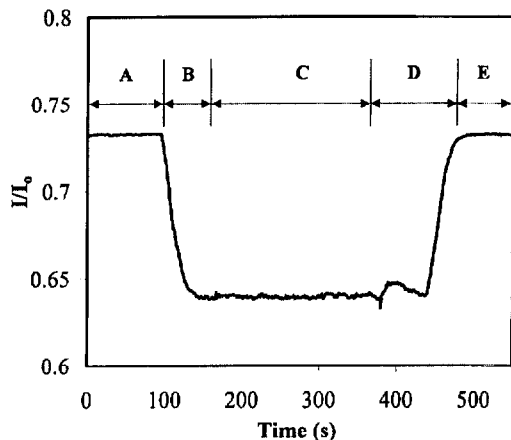


FIG. 2. Transmittance measurements, I/I_0 , performed for laminar acetylene and air diffusion flame soot. Duration A: background monitoring; duration B: soot introduction to transmission cell; duration C: soot collection period; duration D: clean air introduction to transmission cell; duration E: background monitoring.

provides a point measurement of the soot concentration, whereas light extinction provides a line-of-sight average. Near-uniform soot concentration distribution in both the axial and radial directions is required to provide an accurate σ_s . A video camera was used to monitor the scattered light intensity over an 8-cm length near the inlet to assess the uniformity of the smoke in the axial direction along the laser beam. The only region of nonuniformity detected from digitized images was the First 2 mm due to the smoke flow past the purge tube.

A radial gradient could develop from the deposition of particles to the walls as a result of thermophoresis and diffusion. Because the temperature is essentially ambient, thermophoretic transport need not be considered. From a convective diffusion analysis for fully developed pipe flow, one finds that for a flow velocity of 2 cm/s and a diffusion coefficient for a 50-nm sphere, the concentration gradient is less than 0.5% [11]. This is an upper bound estimate because the diffusion coefficient of the agglomerate will be less than that for a single 50-nm sphere. The overall relative uncertainty associated with both radial and axial nonuniformity is 1%.

Experiments were also performed to confirm that particle deposition on the walls does not play an important role in the measurement of σ_s . The reduction in gravimetrically sampled smoke (as a result of the deposition) will produce an artificially large σ_s . Filter measurements were performed at location 1 (before smoke enters the tube) and 2 (after smoke exits the tube). The deposition fraction (difference between measurement at 1 and 2 divided by measurement at 1) was 3% for larger soot particles produced from acetylene flames and 2% with smaller

soot particles generated using ethene flames. Assuming that the deposition occurs uniformly in the tube, the maximum path-averaged difference between optical and gravimetric measurements would be 1.5%.

The light collected by the detector for the transmitted beam includes a small amount of scattered light, which will lower the measured value of σ_s . For an ideal extinction measurement, there would be no scattered light reaching the detector. By performing measurements for acceptance angles ranging from 0.5° to 1.3° , it was found that the scattered light causes the term $\ln(I/I_0)$ to increase by 1%. For the 1.1° aperture used in our experiments, the uncertainty associated with forward-scattered light, I_s , is estimated to be 0.8%.

Table 3 summarizes all of the type B uncertainties. The effects of both the drift in the intensity ratio and the effect of forward-scattered light are included in the uncertainty of $\ln(I_0/I)$. The uncertainty in M_s is obtained by computing the rms of the individual uncertainties for m_s , m_d , V , T , and uniformity factor b . The combined uncertainty of σ_s for the systematic component, u_s , is obtained by the root sum of squares of M_s , L , and $\ln(I_0/I)$ as 2.7% compared with 7.2% for the facility developed by Choi et al. [3].

The most convincing test of the accuracy of the present measurement technique was obtained by comparing the σ_s measurements of monodisperse, polystyrene (PS) spheres with the predicted value. PS spheres have a mean diameter of 496 ± 4 nm, a refractive index of 1.59, and a density of 1.05 g/cm³. The PS sphere aerosol was formed by nebulizing (103-kPa gauge) a suspension of PS spheres in water and then evaporating the water by passage through a diffusion drier. The value of σ_s calculated using Mie theory was 9.57 ± 0.14 m²/g [12]. This is in excellent agreement with the experimentally measured value of 9.19 ± 0.19 m²/g. The range in the experimental values is the standard deviation for repeat measurements while the predicted range includes the variation caused by the ± 4 nm uncertainty in the size of the PS spheres and by the presence of 10% doublets [12]. The type B uncertainty is larger than that for the smoke experiments because of the small values of $\ln(I_0/I)$, about 0.03 compared with about 0.15 for smoke, and the small value of the mass of particles collected, about 0.3 mg compared with at least 1 mg for smoke. The overall type B uncertainty is about 0.05 m²/g for the case of the PS spheres, which is as large as the difference between the measured and predicted value.

In addition to the light extinction measurements, data were also collected on the total scattering using a reciprocal nephelometer [13]. The cosine sensor-photomultiplier tube (PMT) is positioned at the center of the cell and collects light scattered from 1.5° to 178.5° . The reciprocal nephelometer is calibrated by simultaneously measuring the light extinction and

TABLE 3
Reduction in type B uncertainty by using laminar flame/LAOF apparatus

Quantity	SD ^a (x_i)/ x_i , Premixed Choi et al. [3]	SD(x_i)/ x_i , LAOF Present Study	Comments
$\ln(I_0/I)$	0.035	0.017	stabilized laser, steadier soot source, and effect of forward-scattered light-reaching detector
L	0.024	0.005	1-m path length vs. 0.084-m path length
m_s	0.006	0.006	less soot collected but more accurate balance
m_d (smoke deposition effect)	0.0	0.015	wall loss not a factor for premixed apparatus
V	0.020	0.009	improve flow calibration using an automated soap film meter
T	0.021	0.003	soot near ambient temperature
b (uniformity factor)	0.05	0.010	well-mixed soot near ambient temperature
M_{ave}	0.058	0.021	
σ_s	0.072	0.027	

^aSD, standard deviation.

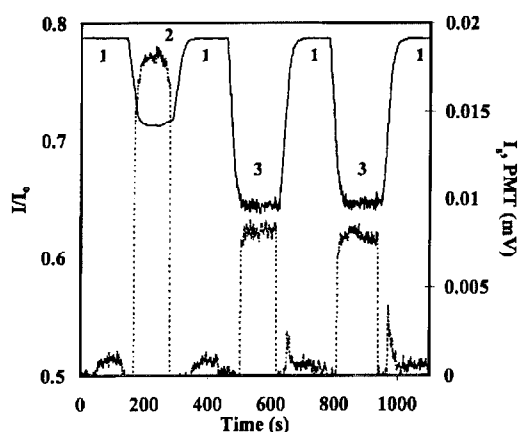


FIG. 3. Simultaneous measurement of transmittance (solid line) and scattering intensity, I_s (dashed line), for laminar acetylene diffusion flame smoke. (1) Corresponds to background measurement. (2) Corresponds to DOP-scattering measurements. (3) Corresponds to smoke-scattering measurements.

the PMT output for a dioctylphthalate (DOP) aerosol. A constant output aerosol is generated by nebulizing a 3%-by-weight solution of DOP dissolved in isopropanol. The isopropanol evaporates leaving a polydisperse aerosol with a mass mean diameter of about $0.5 \mu\text{m}$. The key to the calibration is the fact that DOP does not absorb light so that the extinction cross section is equal to the scattering cross section. The scattering intensity ratio and the PMT output are plotted versus time for background reading, the

DOP aerosol, and soot in Fig. 3. The ratio of the scattering cross section to the extinction cross section is in the range 0.22–0.25 for acetylene compared to 0.19–0.21 for ethene (Table 2). The repeatability of the light-scattering measurement is about $\pm 4\%$ of the mean value.

The primary sphere sizes of the various smokes were measured by transmission electron microscopy (TEM) for samples collected by sedimentation in the middle of the LAOF. The TEM was calibrated using NIST Standard Reference Material 1963, monosize 100-nm polystyrene spheres. The relative increase in primary sphere size of the soot produced by turbulent combustion of acetylene relative to ethene is about 35%, which is close to the result of Köylü and Faeth [19]; however, the magnitude of the values is larger in the present experiments with values of 48 nm (ethene) and 67 nm (acetylene) compared with 32 and 47 nm [19]. The laminar flames produced slightly smaller primary sphere size of 40 nm (ethene) and 61 nm (acetylene). Additional TEM measurements of the soot primary sphere size are planned to help resolve the difference in the size measurements between this study and the study by Köylü and Faeth [19].

Discussion

The mean σ_s measured for acetylene in the turbulent diffusion flame configuration in the present study is $7.80 \text{ m}^2/\text{g}$. This is in excellent agreement with previous measurements of $7.82 \text{ m}^2/\text{g}$ for turbulent acetylene premixed flames [3]. In contrast, Wu et al. [7] report a significantly lower value of 4.5

m^2/g for acetylene turbulent diffusion flame soot. The turbulent diffusion flame experiments in the present study were performed using a burner that is identical to that used by Wu et al. [7]. The reason for the discrepancy is not known, but the reliability of the present results is supported by the absolute calibration and low uncertainties associated with the experimental measurements.

Mulholland and Mountain [14] computed the σ_s of agglomerates with 3 to 1392 primary spheres using a coupled electric and magnetic dipole method (CEMD). The computed σ_s of the largest agglomerate for primary sphere diameters of 42 and 60 nm at a wavelength of 632.8 nm are 6.2 and 6.5 m^2/g . This is for a refractive index of $1.7 + i0.7$ and for a soot density of 1.8 g/cm^3 . Decreasing the real part of the refractive index and increasing the imaginary part results in an increased value of σ_s . It was found that an imaginary part of the refractive index equal to 0.8 was required to obtain a σ_s of 8.0 m^2/g for primary sphere diameter of 42 nm given a real part of the refractive index equal to 1.55. If a larger value for the real part of the refractive index is assumed, then an imaginary value with magnitude greater than 0.8 is required. Dobbins et al. [2] obtained a similar result based on an analytic model for polydisperse fractal agglomerates. The combination of a large imaginary component of the refractive index of about 0.8 and a small real component of about 1.55 is outside the range of measured refractive indices [7,8,15–18].

The high resolution of the LAOF provides the capability to distinguish differences in σ_s due to fuel and burner dependence. It is observed that laminar and turbulent flame conditions alter σ_s for a given fuel by only 3–4%. However, there were much larger differences between the two fuels. The average σ_s , based on 17 acetylene laminar flame experiments on 6 days is $7.55 \pm 0.05 \text{ m}^2/\text{g}$, while the average based on 21 turbulent experiments on 4 days is $7.80 \pm 0.08 \text{ m}^2/\text{g}$. Similarly, the average σ_s , based on 20 ethene laminar flame experiments on 6 days is $8.50 \pm 0.11 \text{ m}^2/\text{g}$, while the average for 19 turbulent experiments on 3 days is $8.79 \pm 0.28 \text{ m}^2/\text{g}$.

The average σ_s for ethene is nearly 12% larger than the value for ethylene, while the primary sphere size for ethene is about 35% smaller than for acetylene. This trend of decreasing σ_s with increasing primary sphere size is inconsistent with the calculated σ_s [14]. For agglomerates with 100 to 1000 primary spheres, the calculated σ_s increases from 5.7 to 6.1 to 6.4 m^2/g as the primary sphere size was increased from 32 to 42 to 60 nm. Based on our measurements of extinction coefficient as a function of acceptance angle, we do not believe that an increase in the forward-scattered light reaching the detector is the cause of this effect. The difference in the observed trends could be explained by differences in the refractive indices of acetylene smoke

and ethene smoke. We believe that a more likely explanation is a shielding effect where the light absorption on one side of the agglomerate reduces the light intensity reaching the other side of the agglomerate. Based on the CEMD calculation, the shielding effect will increase with increasing primary sphere size and increasing agglomerate size such as is observed for the acetylene smoke relative to the ethene smoke.

Conclusions

1. An improved light extinction and filter collection facility has been developed with a total type B (systematic) uncertainty of 2.7% of the mean, compared with 7–10% for previous methods. The combined, expanded uncertainty (at 95% confidence level) is 5.6 and 5.8% of the mean for the laminar and turbulent acetylene smoke and 6.0 and 8.4% of the mean for the laminar and turbulent ethene smoke. For the first time, an absolute calibration using monodisperse polystyrene spheres has been used to validate the reliability of σ_s measurements for smoke.
2. The σ_s values for the smoke from the turbulent acetylene and ethene flames (7.80 and 8.79 m^2/g , respectively) are significantly larger than the values measured in a previous study [7]. However, the present measurements are similar to the values obtained from earlier studies at NIST ranging from 7.3 to 9.5 m^2/g for five fuels [1], 7.8 m^2/g measured for crude oil [2], and 7.8 m^2/g for premixed acetylene [3].
3. Measurements of the σ_s for acetylene and ethene soot indicate that there is a 12% difference based on fuel type and no statistically significant difference based on flame conditions (either laminar or turbulent cases). The difference measured for the two fuels may be caused by a shielding effect that is dependent on primary sphere size and the number of spheres comprising an agglomerate.
4. The measured values of σ_s in the present study appear to be inconsistent with the range of refractive index values widely used in the combustion community [7,8,15–18].

Acknowledgment

The σ_s of a polystyrene sphere doublet was computed by Egon Marx from the Manufacturing Engineering Lab at NIST.

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COMMENTS

Daniel E. Rosner, Yale University, USA. Evidently, large fire at atmospheric pressure can produce soot aggregate with characteristic dimensions of the order of 1000 μ m, containing spherules with diameters over 100 nm. When the host gas temperatures are not very high this corresponds to soot aggregates much larger than the gas mean free path, with interesting (but, as yet, little studied) consequences for the associated transport properties. Motivated by comparable effects expected at high pressure, we have been studying the thermophoretic properties of such factal-like aggregates, that is, how rapidly they would drift in a temperature gradient. Our conclusion [1,2], potentially important for predicting soot deposition rates from fires, is that their thermophoretic diffusivity will remain comparable to that in the free-molecule limit because of their low effective thermal conductivity. The coagulation rate of such radiation-cooled aggregates can also be enhanced by thermophoretic effects [3]. Have you seen any evidence of these

small Knudsen number effects for such giant aggregates in fire environments at 1 atm?

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Author's Reply. Even for the laminar acetylene flame, we observed individual agglomerates with a characteristic dimension on the order of 1000 μ m. We do not have any data on the thermophoretic diffusivity of such large agglomerates to compare with the Rosner-Khalil prediction of free-molecule behavior.